

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization  
International Bureau



(43) International Publication Date  
23 October 2003 (23.10.2003)

PCT

(10) International Publication Number  
WO 03/087258 A1

(51) International Patent Classification<sup>7</sup>: C09K 11/06,  
H05B 33/14, 33/22

(21) International Application Number: PCT/JP03/04483

(22) International Filing Date: 9 April 2003 (09.04.2003)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:  
2002-109939 12 April 2002 (12.04.2002) JP

(71) Applicant (for all designated States except US): CANON  
KABUSHIKI KAISHA [JP/JP]; 3-30-2, Shimomaruko,  
Ohta-ku, Tokyo 146-8501 (JP).

(72) Inventors; and

(75) Inventors/Applicants (for US only): YAMADA, Naoki  
[JP/JP]; c/o CANON KABUSHIKI KAISHA, 3-30-2,

Shimomaruko, Ohta-ku, Tokyo 146-8501 (JP). UENO,  
Kazunori [JP/JP]; c/o CANON KABUSHIKI KAISHA,  
3-30-2, Shimomaruko, Ohta-ku, Tokyo 146-8501 (JP).

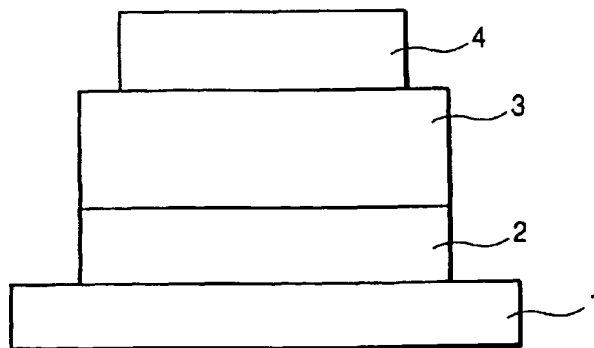
(74) Agents: OKABE, Masao et al.; 602, Fuji Bldg., 2-3,  
Marunouchi 3-chome, Chiyoda-ku, Tokyo 100-0005 (JP).

(81) Designated States (national): AE, AG, AL, AM, AT, AU,  
AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU,  
CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH,  
GM, HR, HU, ID, IL, IN, IS, KE, KG, KP, KR, KZ, LC,  
LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW,  
MX, MZ, NI, NO, NZ, OM, PH, PL, PT, RO, RU, SC, SD,  
SE, SG, SK, SL, TJ, TM, TN, TR, TT, TZ, UA, UG, US,  
UZ, VC, VN, YU, ZA, ZM, ZW.

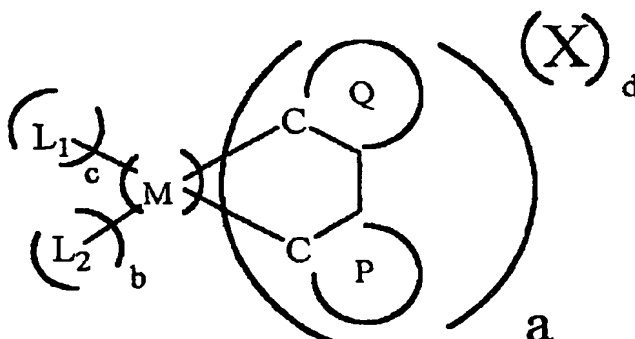
(84) Designated States (regional): ARIPO patent (GH, GM,  
KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW),  
Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),  
European patent (AT, BE, BG, CH, CY, CZ, DE, DK, EE,

[Continued on next page]

(54) Title: ORGANIC LIGHT-EMITTING DEVICE



(57) Abstract: An organic light-emitting device that exhibits a light emission of extremely high efficiency, high brightness, and long lifetime is provided. An organic light-emitting device includes a pair of electrodes including an anode and a cathode, and one or more layers containing an organic compound, the layers being sandwiched between the pair of the electrodes, characterized in that at least one layer of the layers containing an organic compound contains a compound represented by the following general formula (1).



(1)



ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PT, RO,  
SE, SI, SK, TR), OAPI patent (BF, BJ, CF, CG, CI, CM,  
GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

*For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

**Published:**

— *with international search report*

## DESCRIPTION

## ORGANIC LIGHT-EMITTING DEVICE

## 5 TECHNICAL FIELD

The present invention relates to an organic light-emitting device, and more specifically to a device that emits light by applying an electric field to a thin film containing an organic compound.

10

## BACKGROUND ART

Organic light-emitting devices are luminescent devices utilizing light that is emitted when excitons of fluorescent organic compounds move back to the ground state, the excitons being formed by sandwiching a thin film that contains the fluorescent compounds between an anode and a cathode and injecting electrons and holes (positive holes) from the respective electrodes.

20 A study by Eastman Kodak Company in 1987 (Appl. Phys. Lett. 51, 913 (1987)) reports that a light emission of about  $1000 \text{ cd/m}^2$  was observed under an applied voltage of about 10 V. In the study, the device had a function-separated type bilayer structure and used ITO as the anode, a magnesium-silver alloy as the cathode, an aluminum quinolinol complex as an electron-transporting material and a

25

light-emitting material, and a triphenylamine derivative as a hole-transporting material. Related patent documents include US Patents No. 4539507, No. 4720432, and No. 4885211, etc.

5           Moreover, various light emissions in a range of from ultraviolet to infrared can be accomplished by using various different fluorescent organic compounds. Recently, a lot of studies on various compounds have been conducted actively. For instance, those studies  
10           are disclosed in US Patents No. 5151629, No. 5409783, No. 5382477, No. 5130603, No. 6093864, No. 5227252, Japanese Patent Applications Laid-Open No. 5-202356, No. 9-202878, and No. 9-227576.

          In addition to the organic light-emitting  
15           devices using small molecular materials as described above, the organic light-emitting devices using conjugated polymers have been reported by the group of Cambridge University (Nature, 347, 539 (1990)). This report discloses that a film of  
20           polyphenylenevinylene (PPV) is formed by coating method and it is surely observed that a single layer of the film emitted light. Related patent documents on organic light-emitting devices using conjugated polymers include US Patents No. 5247190, No. 5514878,  
25           No. 5672678, No. 5317169, No. 5726457, and Japanese Patent Application Laid-Open No. 5-247460, etc.

          Thus the remarkable progresses have been made

in organic light-emitting devices, so that the recent luminescent devices are characterized by high brightness even when a lower voltage is applied, diversity in emission wavelengths, and rapid response, and thin and lightweight luminescent devices can be obtained, which suggests a wide range of possibilities of the device being utilized for various uses.

However, under the present circumstances, light emissions exhibiting higher brightness or higher conversion efficiency are required. Furthermore, there are still many problems in regard to durability such as change with elapse of time due to prolonged use and deterioration or the like, by moisture and the atmospheric gas containing oxygen. Moreover, in view of application to a full-color display etc., blue, green, and red light emissions having higher color purities are needed, and this problem has not been solved sufficiently yet.

#### SUMMARY OF THE INVENTION

The present invention has been made to solve the above-mentioned problems of the conventional technologies, and an object of the present invention is to provide an organic light-emitting device that exhibits a light emission of extremely high efficiency, high brightness, and long lifetime.

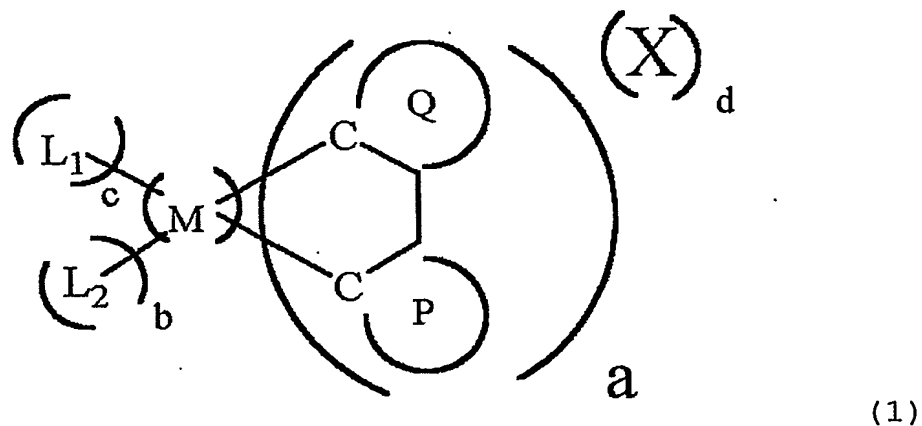
Furthermore, another object of the present invention is to provide an organic light-emitting device that shows various emission wavelengths and various emission hues and that is extremely durable.

5        Moreover, another object of the present invention is to provide an organic light-emitting device that can not only be manufactured easily but also produced at a relatively low cost.

10        The inventors of the present invention made intensive investigations to solve the above-mentioned problems. As a result, they have found out that an organic light-emitting device exhibiting a light emission of higher efficiency and higher brightness can be produced by use of an organic light-emitting  
15        device including a pair of electrodes including an anode and a cathode, and one or more layers containing an organic compound, the layers being sandwiched between the pair of the electrodes, in which at least one layer of the layers containing an  
20        organic compound contains a specific compound, thereby completing the present invention.

      According to the present invention, there is provided an organic light-emitting device including a pair of electrodes including an anode and a cathode,  
25        and one or more layers containing an organic compound, the layers being sandwiched between the pair of the electrodes, in which at least one layer of the layers

containing an organic compound contains a compound represented by the following general formula (1).



(In the formula, M is a metal atom, preferably an  
 5 iridium atom or a platinum atom; P and Q each are a  
 substituted or unsubstituted aromatic hydrocarbon  
 group or a substituted or unsubstituted aromatic  
 heterocyclic group; and P and Q constitute a chelate  
 ligand; P and Q may form a condensed ring with each  
 10 other; L<sub>1</sub> and L<sub>2</sub> each are a ligand or an oxygen atom  
 that binds to M by a double bond; L<sub>1</sub> and L<sub>2</sub> may form a  
 condensed ring with each other to become a two-  
 coordinate chelate ligand; X represents a counter ion  
 of an anion or a cation; a is an integer of 1 to 3; b  
 15 and c each are an integer of 0 to 4; and d is an  
 integer of 0 to 3, provided that P, Q, L<sub>1</sub>, L<sub>2</sub>, and X  
 each may be the same or different if there are more  
 than one of these).

In the organic light-emitting device of the  
 20 present invention, the metal atom M preferably is a

five-coordinate 16-electron bonding system or a six-coordinate 18-electron bonding system with  $L_1$  and  $L_2$  and with a chelate ligand comprising P and Q.

## 5 BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a sectional view showing an example of an organic light-emitting device of the present invention.

Fig. 2 is a sectional view showing another  
10 example of an organic light-emitting device of the present invention.

Fig. 3 is a sectional view showing still another example of an organic light-emitting device of the present invention.

15

## BEST MODES FOR CARRYING OUT THE INVENTION

Hereinafter, the present invention will be explained in detail.

First, the compounds represented by the above-  
20 mentioned general formula (1) for use in the present invention will be explained.

Examples of the metal M in the general formula (1) include Pd(II), Pt(II), Ru(II)CO, Zn(II), Mg(II), Sn(IV), Pb(II), Al, Cd, Si(IV), Ge(IV), Ba, Sr, Be,  
25 Sc(III), Ti(IV), Zr(IV), Hf(IV), Nb(V), Ta(V), Co(III), Rh(III), Ir(III), Ir(IV), Ni(II), VO, Sn(II), Ru(II), Ru(III), Cu(II), Ag, Co(II), Ca, Hg, Mn(II),



Fe(II), Fe(III), and the like, but are not limited to the above-mentioned metal and a general metal atom may be used.

Further, specific examples of P and Q that  
5 constitute a chelate ligand will be shown hereinbelow.

The substituted or unsubstituted aromatic hydrocarbon group includes a phenyl group, a 4-methylphenyl group, a 4-ethylphenyl group, a 3-chlorophenyl group, a 3,5-dimethylphenyl group, a  
10 triphenylamino group, a biphenyl group, a terphenyl group, a naphthyl group, an anthryl group, a phenanthryl group, a pyrenyl group, a tolyl group, a tosyl group, a halogen-substituted naphthyl group, and the like.

15 The substituted or unsubstituted aromatic heterocyclic group includes: five-membered heterocyclic rings such as a thienyl group, a furyl group, a pyrrolyl group, an imidazolyl group, a pyrazolyl group, an isothiazolyl group, and an  
20 isoxazolyl group; six-membered heterocyclic rings such as a pyranyl group, a pyridyl group, a pyrazinyl group, a pyrimidinyl group, and a pyridazinyl group; and a bipyridyl group, a methylpyridyl group, a terthienyl group, a propylthienyl group, an  
25 isobenzofuranyl group, an indoliziny group, an isoindolyl group, an indolyl group, an indazolyl group, a purinyl group, an indolinyl group, an

isoindolinylyl group, a chromenyl group, a quinolizinylyl group, an isoquinolyl group, a quinolyl group, a phthalazinylyl group, a naphthylidinylyl group, a quinazolinyl group, a carbazolyl group, an N-ethylcarbazolyl group, a thianthrenyl group, phenanthridinylyl group, a perimidinylyl group, etc.

The condensed ring that P and Q forms include a benzo ring, a naphtho ring, an anthra ring, an acenaphtho ring, and the like. It may also include heterocyclic rings such as those whose names begin with furo-, imidazo-, pyrido-, quino-, thieno-, and the like.

In addition, the substituents that P and Q may have include but are of course not limited to: alkyl groups or aralkyl groups such as a methyl group, an ethyl group, an n-propyl group, an iso-propyl group, a tert-butyl group, an octyl group, a benzyl group, and a phenethyl group; alkenyl groups such as a vinyl group, an allyl group (a 2-propenyl group), a 1-propenyl group, an iso-propenyl group, and a 2-butenyl group; alkoxy groups such as a methoxy group, an ethoxy group, a propoxy group, a 2-ethyloctyloxy group, a phenoxy group, a 4-butylphenoxy group, and a benzyloxy group; carbonyl groups such as an acetyl group, a propionyl group, an isobutyryl group, a methacryloyl group, a benzoyl group, a naphthoyl group, an anthroyl group, and a toluoyl group; amino

groups such as a methylamino group, an ethylamino group, a dimethylamino group, a diethylamino group, a methylethylamino group, a benzylamino group, a methylbenzylamino group, an anilino group, a  
5 diphenylamino group, a phenyltolylamino group, and a ditolylamino group; a halogen; an azo group; those aryl groups, heterocyclic groups, or condensated rings described above; and the like.

Also, specific examples of the ligands  $L_1$  and  $L_2$   
10 include alkoxy groups such as a methoxy group, an ethoxy group, a propoxy group, a 2-ethyloctyloxy group, a phenoxy group, a 4-butylphenoxy group, and a benzyloxy group.

In addition, it may include aryl groups such as  
15 a phenyl group, a 4-methylphenyl group, a 4-ethylphenyl group, a 3-chlorophenyl group, a 3,5-dimethylphenyl group, a triphenylamino group, a biphenyl group, a terphenyl group, a naphthyl group, an anthryl group, a phenanthryl group, a pyrenyl  
20 group, a tolyl group, a tosyl group, a halogen-substituted naphthyl group, and the like.

Further, it may include: five-membered heterocyclic rings such as a thienyl group, a furyl group, a pyrrolyl group, an imidazolyl group, a  
25 pyrazolyl group, an isothiazolyl group, and an isoxazolyl group; six-membered heterocyclic rings such as a pyranyl group, a pyridyl group, a pyrazinyl

group; a pyrimidinyl group, and a pyridazinyl group; and heterocyclic groups such as a bipyridyl group, a methylpyridyl group, a terthienyl group, a propylthienyl group, an isobenzofuranyl group, an indoliziny 5 l group, an isoindolyl group, an indolyl group, an indazolyl group, a purinyl group, an indolinyl group, an isoindolinyl group, a chromenyl group, a quinoliziny 10 l group, an isoquinolyl group, a quinolyl group, a phthalazinyl group, a naphthylidinyl group, a quinazolinyl group, a carbazolyl group, an N-ethylcarbazolyl group, a thianthrenyl group, phenanthridinyl group, a perimidinyl group, etc.

Furthermore, it may include phosphino groups such as a triphenylphosphino group, a trimethylphosphino group, and a triethylphosphino group.

Additionally, it may also include amino groups such as a methylamino group, an ethylamino group, a dimethylamino group, a diethylamino group, a methylethylamino group, a benzylamino group, a methylbenzylamino group, an anilino group, a diphenylamino group, a phenyltolylamino group, and a ditolylamino group.

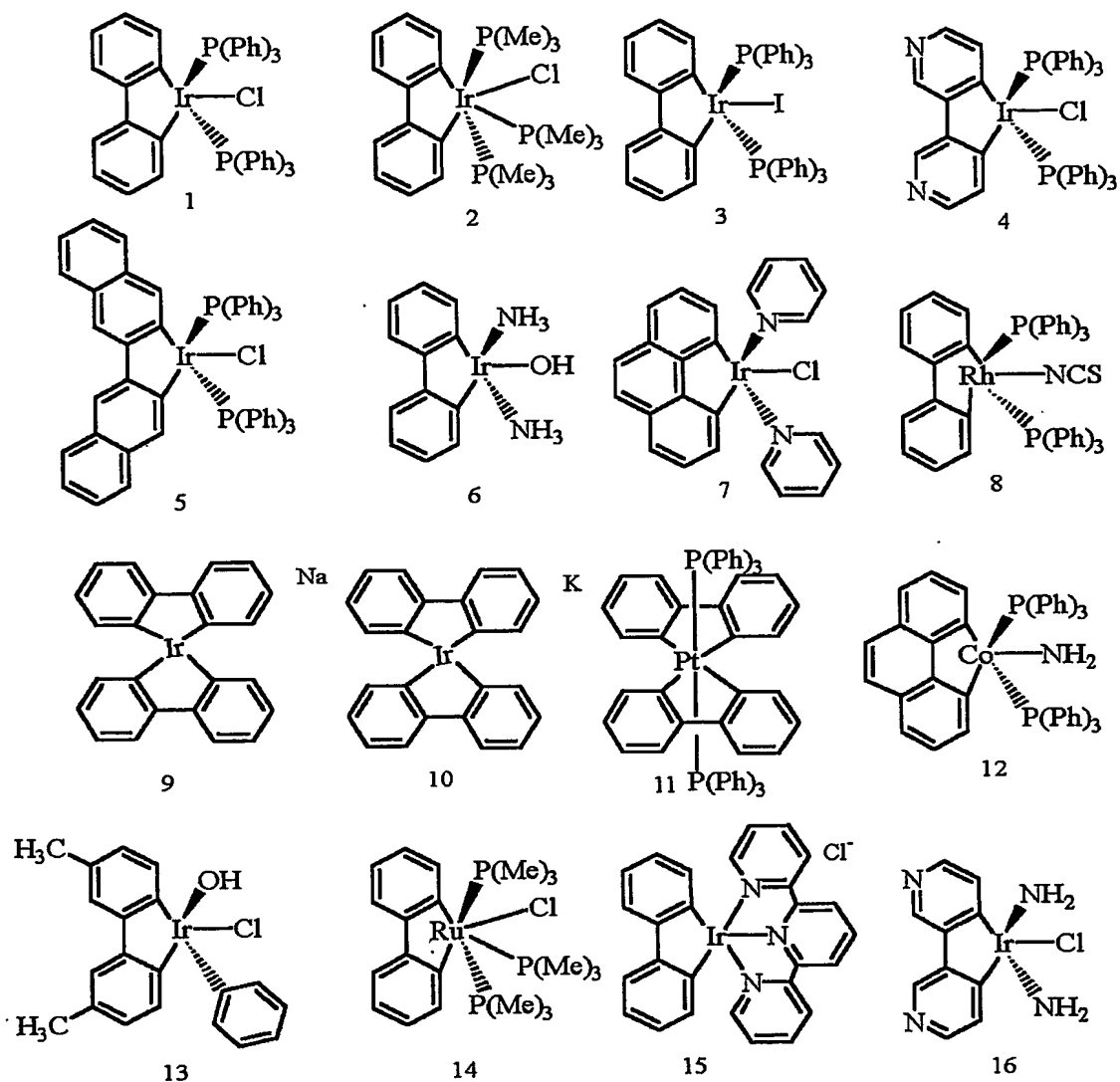
Moreover, azo groups that were formed by optionally combining the above-mentioned substituents may be used.

For the condensed rings that  $L_1$  and  $L_2$  form, a benzo ring, a naphtho ring, an anthra rings, an acenaphto ring, and the like may be used. It may also include heterocyclic rings such as those whose  
5 names begin with furo-, imidazo-, pyrido-, quino-, thieno-, and the like.

Also,  $L_1$  and  $L_2$  may have substituents, and those substituents include, but of course are not limited to, the above-mentioned alkoxy groups, heterocyclic  
10 groups, phosphino groups, amino groups, halogen groups, azo groups, condensed rings, and the like.

As the counter ion X, there may be mentioned anionic ones in which case examples thereof include  $F^-$ ,  $Cl^-$ ,  $Br^-$ ,  $O_2^-$ ,  $(O_2)^{2-}$ ,  $(O_2)^-$ ,  $(OH)^-$ ,  $(SH)^-$ ,  $(SO_4)^{2-}$ ,  
15  $NH_2^-$ ,  $N^3^-$ ,  $(CN)^-$ ,  $(NCO)^-$ ,  $(NCS)^-$ ,  $(NO_2)^-$ ,  $(OAc)^-$ ,  $(ClO_4)^-$ ,  $(SbO_4)^-$ ,  $(Im)^-$ ,  $(BF_4)^-$ , etc., and cationic ones in which case examples thereof include  $K^+$ ,  $Na^+$ ,  $NH_4^+$ ,  $Li^+$ , etc., but are not limited to the above-mentioned ion and general counter ions may be used  
20 therefor.

Next, representative examples of the compound represented by the general formula (1) will be mentioned. However, the compounds are not limited to the following exemplified compounds.



Next, the organic light-emitting devices of the present invention will be explained with reference to the figures.

Figure 1 is a sectional view showing an example of an organic light-emitting device of the present invention. Figure 1 shows a structure having a substrate 1 on which an anode 2, an emission layer 3,

and a cathode 4 are provided in this order. This light-emitting device is useful in the case of a device independently having hole-transporting ability, electron-transporting ability, and light-emitting ability in itself or the case of a device using a mixture of the compounds having the respective characteristics.

Figure 2 is a sectional view showing another example of an organic light-emitting device of the present invention. Figure 2 shows a structure having a substrate 1 on which an anode 2, a hole-transporting layer 5, an electron-transporting layer 6, and a cathode 4 are provided in this order. This structure is useful in the case of using either a hole-transporting material or an electron-transporting material or a material that have both hole-transporting ability and electron-transporting ability in the respective layers and using a light-emitting material in combination with a mere hole-transporting material or a mere electron-transporting material having no light-emitting ability. In addition, this structure contains a light-emitting layer including either a hole-transporting layer 5 or an electron-transporting layer 6.

Figure 3 is a sectional view showing another example of an organic light-emitting device of the present invention. Figure 3 shows a structure having

a substrate 1 on which an anode 2, a hole-transporting layer 5, a light-emitting layer 3, an electron-transporting layer 6, and a cathode 4 are provided in this order. In this case, carrier transfer function and light emission function are separated. This case is employed appropriately in combination with the respective compounds having hole-transporting ability, electron-transporting ability, and light-emitting ability, respectively. This case, therefore, enables not only extremely increased freedom of selection of materials but also diversity of luminescent hue because various compounds that have different emission wavelengths from each other can be used. Moreover, it is also possible to improve emission efficiency because the respective carriers or excitons are enclosed effectively in the central light-emitting layer 3.

However, Figures 1 to 3 merely show very basic device structures, to which structures of the organic light-emitting devices using the compounds of the present invention are not limited. For example, various layer structures can be prepared, such as by providing an insulating layer in the interface between an electrode and an organic layer, providing an adhesion layer or an interfering layer, and preparing a hole-transporting layer including two layers having different ionization potentials to each



other, etc.

The compounds represented by the general formula (1) used in the present invention are excellent in electron-injecting ability, electron-transporting ability, and light-emitting ability compared with conventional compounds, and can be used in any form as in Figures 1 to 3.

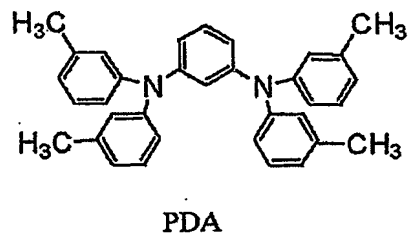
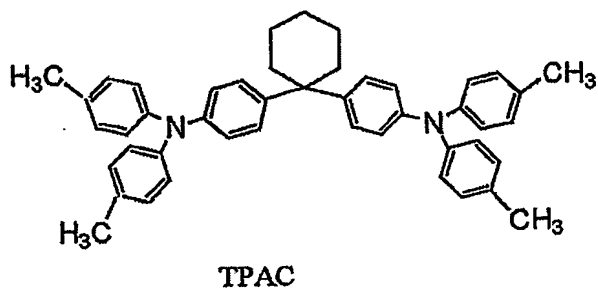
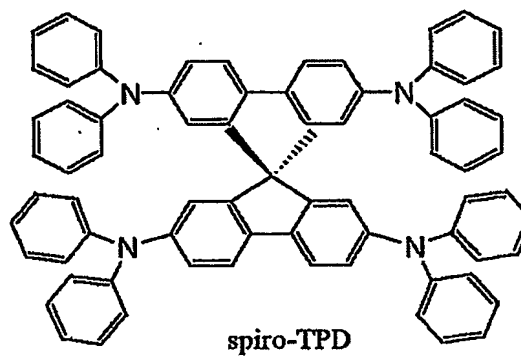
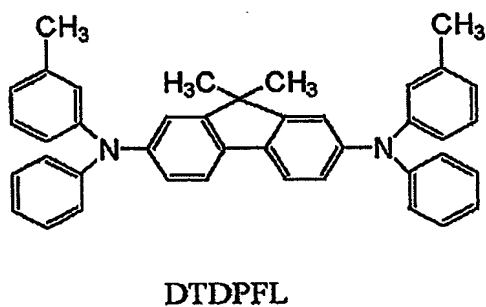
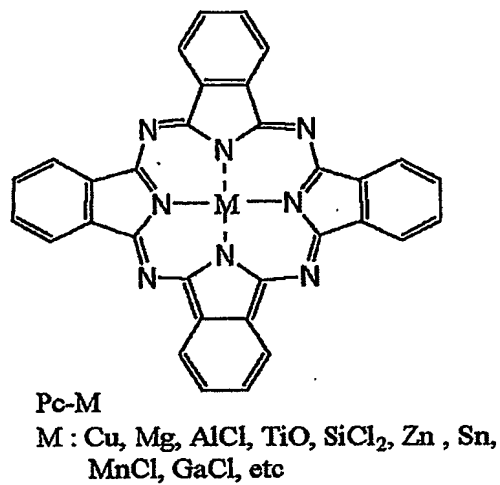
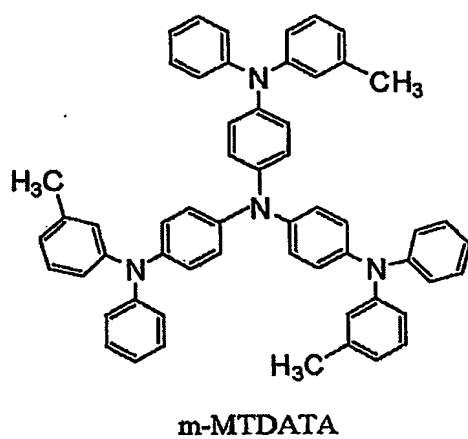
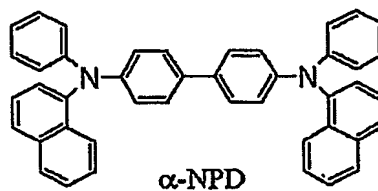
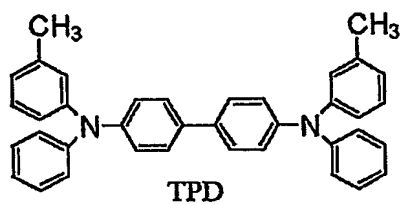
In particular, an organic layer containing a compound represented by the general formula (1) of the present invention is useful as an electron-transporting layer and also as a light-emitting layer.

In the organic light-emitting devices of the present invention, a compound represented by the general formula (1) is made into an organic layer between an anode 2 and a cathode 4 by vacuum vapor deposition method or solution coating method. The thickness of the organic layer is less than 10  $\mu\text{m}$ , and is preferably thinned to a thickness of 0.5  $\mu\text{m}$  or less, more preferably 0.01 to 0.5  $\mu\text{m}$ .

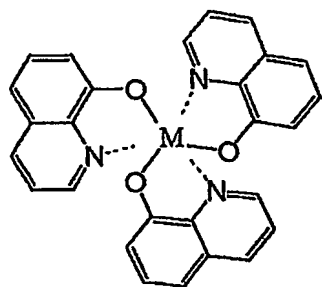
In the present invention, a compound represented by the above-mentioned general formula (1) is used as a component for constituting an electron-transporting layer or a light-emitting layer but additionally, hole-transporting compounds, light-emitting compounds, light-emitting layer matrix compounds, electron-transporting compounds, charge-transporting polymer materials, and light-emitting

polymer materials that have already been known (such as the following compounds) can be used together if necessary. However, such compounds for use in the present invention are not limited to these compounds.

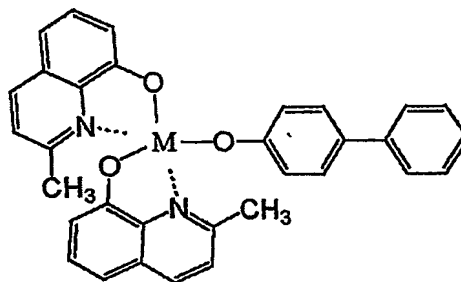
## Hole-transporting compounds



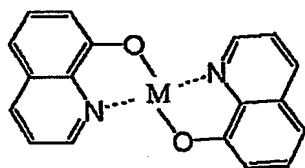
## Electron-transporting light-emitting materials



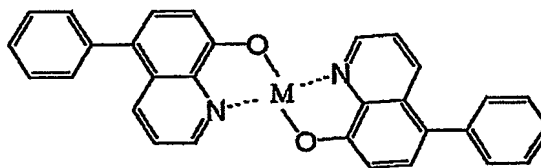
M : Al , Ga



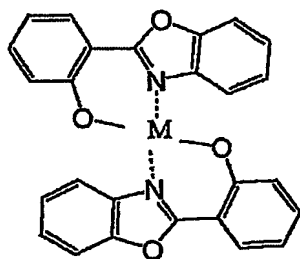
M : Al , Ga



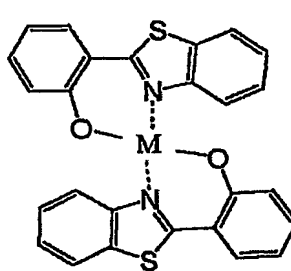
M : Zn , Mg , Be



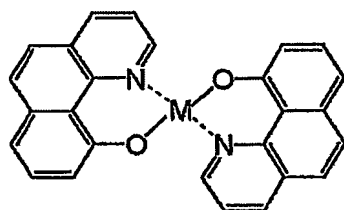
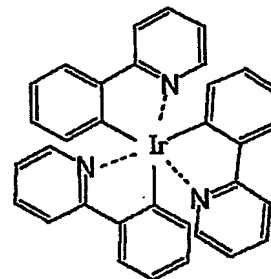
M : Zn , Mg , Be



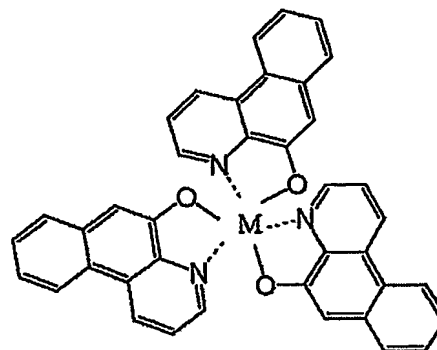
M : Zn , Mg , Be



M : Zn , Mg , Be

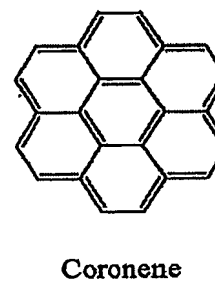
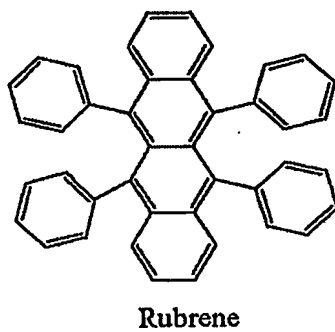
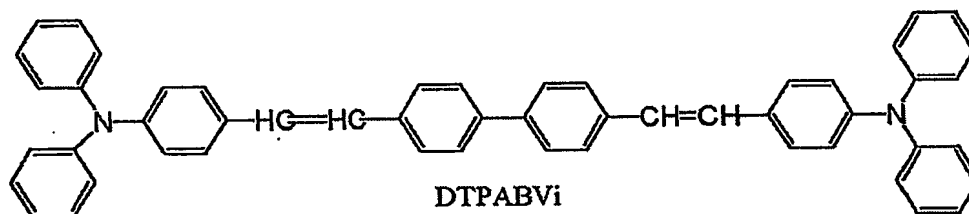
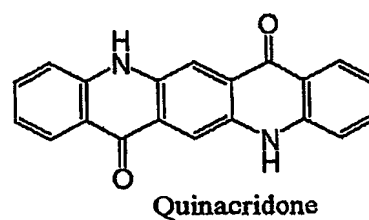
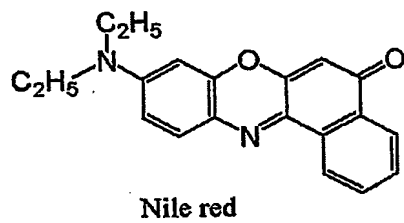
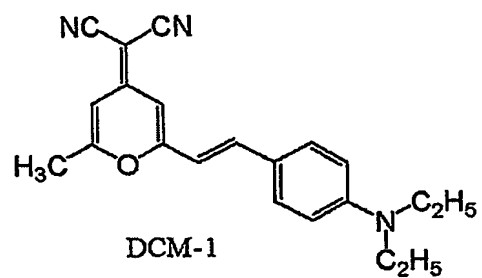
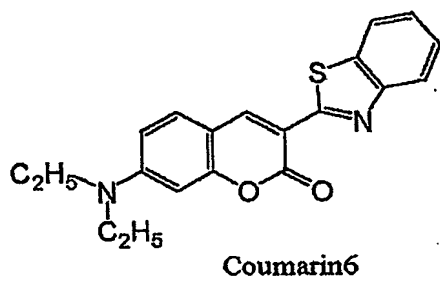


M : Zn , Mg , Be

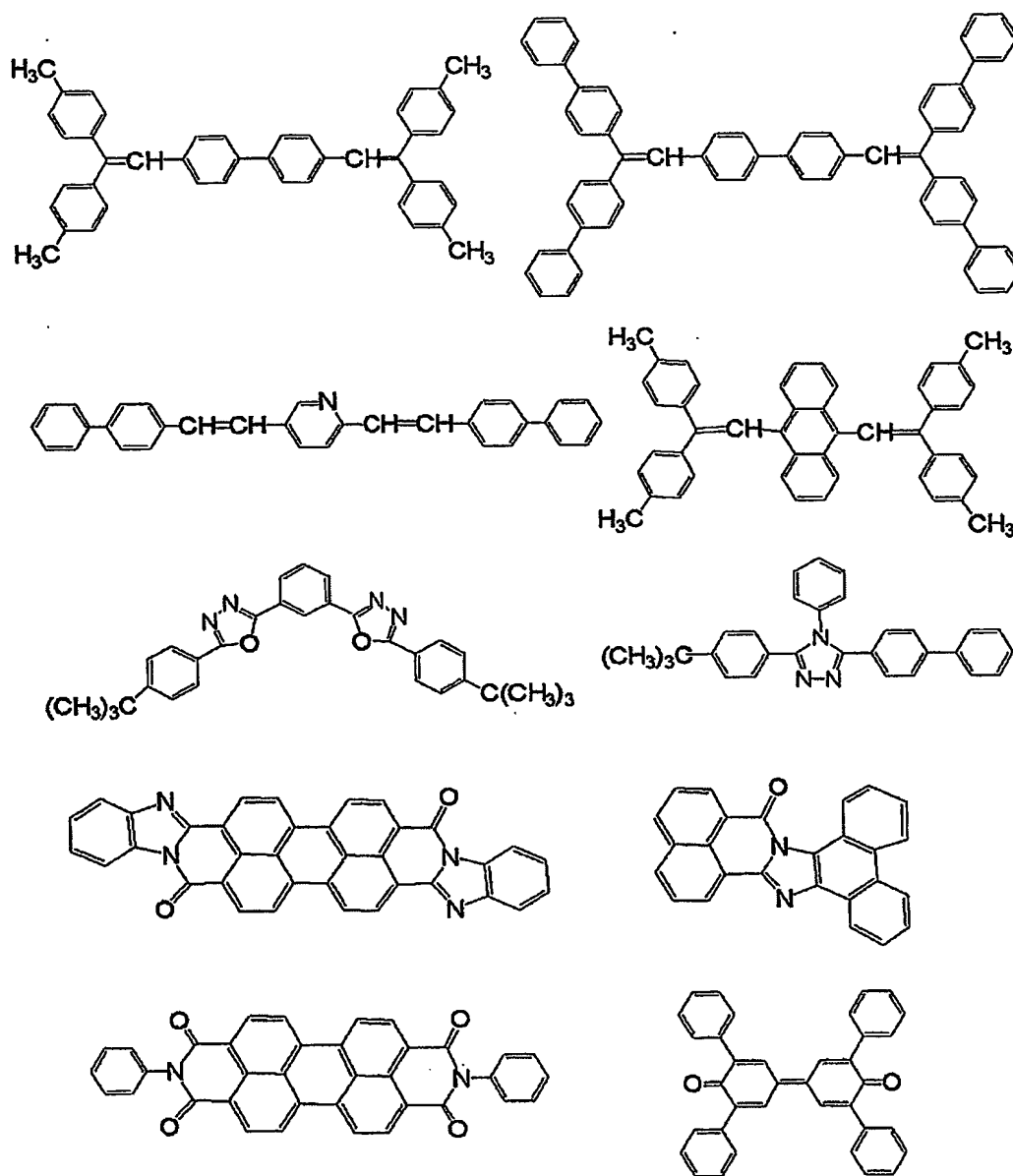


M : Al , Ga

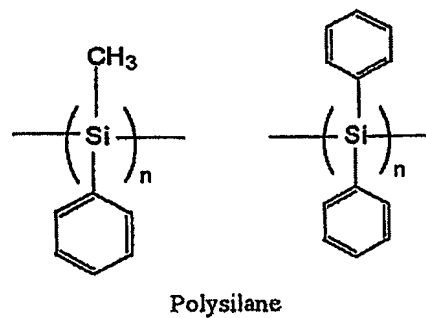
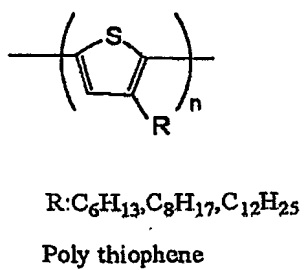
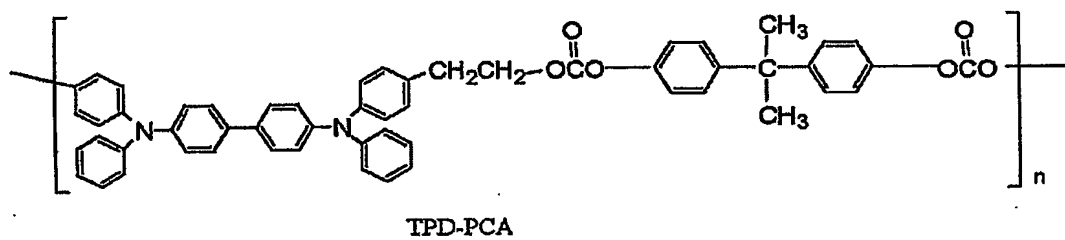
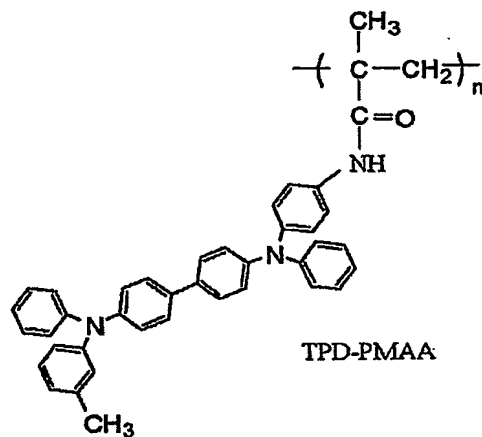
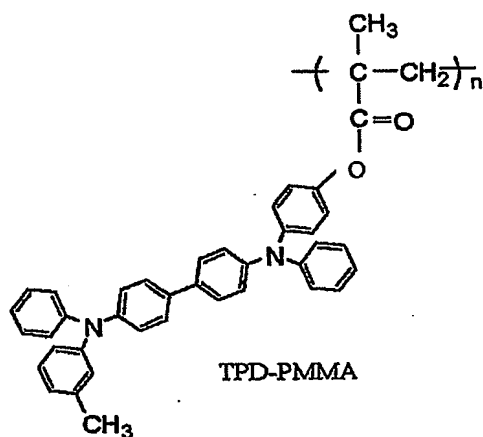
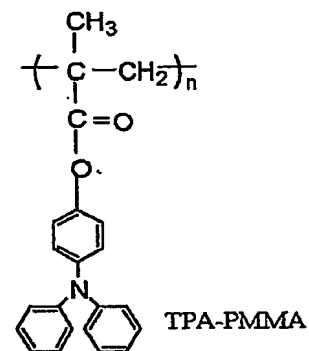
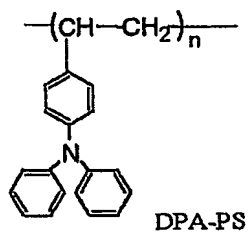
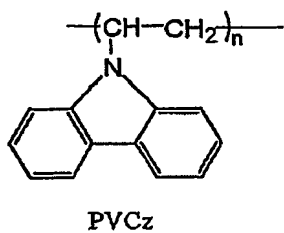
## Light-emitting materials



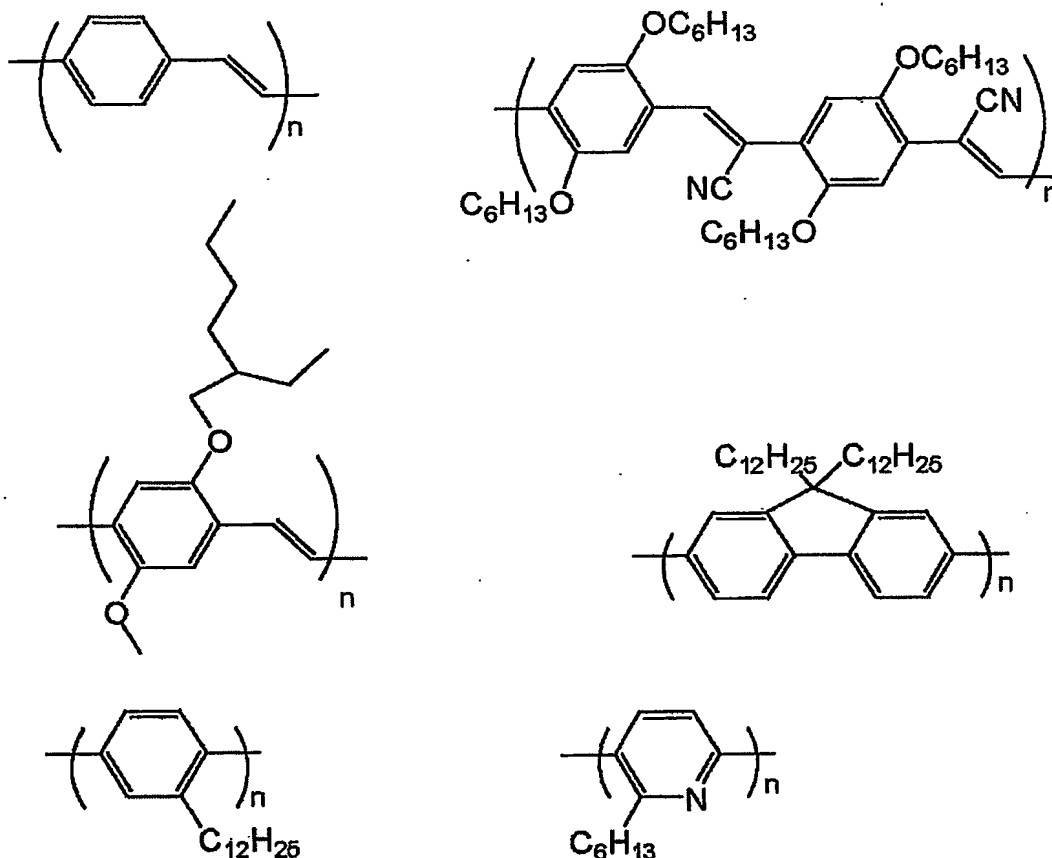
## Light-emitting layer matrix materials and electron-transporting materials



## Polymeric hole-transporting materials



Polymeric light-emitting materials and charge-transporting materials



5            In the organic light-emitting devices of the present invention, a layer containing the compounds represented by the general formula (1) and a layer containing other organic compounds can be made into a thin film generally by vacuum vapor deposition method

10   or by coating method after the compounds are dissolved in an appropriate solvent. In particular,



when a film is prepared by coating method, the compounds can be combined with an appropriate binding resin to form a film.

The above-mentioned binding resins can be  
5 selected from a wide range of binding resins, such as polyvinylcarbazole resin, polycarbonate resin, polyester resin, polyarylate resin, polystyrene resin, acrylic resin, methacrylic resin, butyral resin, poly(vinyl acetal) resin, diallyl phthalate resin,  
10 phenol resin, epoxy resin, silicone resin, polysulfone resin, urea resin, etc., to which the resins are not limited. Moreover, these resins may be used singly, or one or more resins may be mixed with one another to be used as a copolymer.

15 The anode materials are preferably selected from materials having the greatest work function possible. For example, metal simple substances such as gold, platinum, nickel, palladium, cobalt, selenium, and vanadium, or alloys thereof, metal  
20 oxides such as tin oxide, zinc oxide, indium-tin oxide (ITO), and indium-zinc oxide can be used. Furthermore, conductive polymers such as polyaniline, polypyrrole, polythiophene, and polyphenylene sulfide also can be used. These electrode materials may be  
25 used singly or in combination with one another.

On the other hand, the cathode materials are preferably selected from materials having a small

work function. For example, metal simple substances such as lithium, sodium, potassium, calcium, magnesium, aluminum, indium, silver, lead, tin, and chromium, or alloys of some of these can be used.

- 5 Metal oxides such as indium-tin oxide (ITO) also can be used. Additionally, the structure of the cathode may be a single layer or multiple layers.

The substrates for use in the present invention are not particularly limited. For example, opaque  
10 substrates such as metal substrates and ceramic substrates and transparent substrates such as glass, quartz, and plastic sheet are used. Moreover, emitted lights can be controlled by the use of a color filter film, a fluorescent color conversion  
15 filter film, a dielectric reflection film, etc. on a substrate.

Besides, a protection layer or a seal layer may be provided in a prepared device for the purpose of preventing the device from contact with oxygen,  
20 moisture, or the like. Examples of the materials for use in the protection layer include inorganic material films such as diamond thin film, metal oxides, and metal nitrides, polymer films such as fluororesin, poly(p-xylene), polyethylene, silicone  
25 resin, and polystyrene resin, light-setting resin, etc. Furthermore, the prepared devices themselves can be packaged with appropriate seal resins by means

of covering the devices with glass, gas-impermeable films, metals, etc.

#### EXAMPLES

Hereinafter, the present invention will be specifically explained with reference to the following examples, but the present invention is not limited to these examples.

##### (EXAMPLE 1)

A device having a structure as shown in Figure 1 was produced.

A transparent conductive supporting substrate was used which was produced by forming a film of indium-tin oxide (ITO) having a film thickness of 120 nm as an anode 2 on a glass substrate as a substrate 1 by means of sputtering method. This substrate was cleaned ultrasonically with acetone and isopropyl alcohol (IPA) in this order. After boiling cleaning with IPA, the substrate was dried. The substrate was further cleaned with UV/ozone and then was used as a transparent conducting supporting substrate.

A solution was prepared by dissolving 0.050 g of the compound shown as the above-exemplified compound No.1 and 1.00 g of polyvinylcarbazole in 75 ml of tetrahydrofuran. Using this solution, an organic film layer (light-emitting layer 3) having a thickness of 120 nm was formed on a transparent conductive supporting substrate by means of spin coat

method (2000 rpm).

Next, using a deposition material composed of aluminum and lithium (having a lithium concentration of 1 atom%), a metal film layer (cathode 4) having a thickness of 150 nm was formed on the above-mentioned organic layer by means of vacuum vapor deposition method. The film formation was performed under the conditions where the degree of vacuum at the time of vapor deposition was  $1.0 \times 10^{-4}$  Pa, and the film-forming rate was 1.0 to 1.2 nm per second.

Using an ITO electrode as a positive electrode and an Al-Li electrode as a negative electrode, a direct current voltage of 8 V was applied to the device thus obtained, so that flow of an electric current having a current density of  $2.4 \text{ mA/cm}^2$  was observed and a red-light emission having an initial brightness of  $150 \text{ cd/m}^2$  was observed.

(EXAMPLE 2)

A device having a structure as shown in Figure 2 was produced.

A hole-transporting layer 5 was formed by making  $\alpha$ -NPD into a thin film having a thickness of 70 nm on a transparent conductive supporting substrate similar to that in Example 1 by means of vacuum vapor deposition method. Subsequently, an electron-transporting layer 6 was formed by making the exemplified compound No. 15 into a thin film

having a thickness of 50 nm by means of vacuum vapor deposition method. The film formation was performed under the conditions where the degree of vacuum at the time of vapor deposition was  $1.0 \times 10^{-4}$  Pa, and  
5 the film-forming rate was 0.2 to 0.3 nm per second.

Next, a cathode 4 was formed on the above-mentioned organic layer in the same manner as in Example 1.

Using an ITO electrode as a positive electrode  
10 and an Al-Li electrode as a negative electrode, a direct current voltage of 8 V was applied to the device thus obtained, so that flow of an electric current having a current density of  $5.1 \text{ mA/cm}^2$  was observed and a red-light emission having an initial  
15 brightness of  $330 \text{ cd/m}^2$  was observed.

(EXAMPLE 3)

A device having a structure as shown in Figure 3 was produced.

A hole-transporting layer 5 was formed by  
20 making TPD into a thin film having a thickness of 70 nm on a transparent conductive supporting substrate similar to that in Example 1 by means of vacuum vapor deposition method. A light-emitting layer 3 was formed by making the exemplified compound No. 1 into  
25 a thin film having a thickness of 40 nm by means of vacuum vapor deposition method. An electron-transporting layer 6 was formed by making aluminum

trisquinolinol into a thin film having a thickness of 25 nm by means of vacuum vapor deposition method. The film formation was performed under the conditions where the degree of vacuum at the time of vapor  
5 deposition was  $1.0 \times 10^{-4}$  Pa, and the film-forming rate was 0.2 to 0.3 nm per second.

Next, a cathode 4 was formed on the above-mentioned organic layer in the same manner as in Example 1.

10 Using an ITO electrode as a positive electrode and an Al-Li electrode as a negative electrode, a direct current voltage of 8 V was applied to the device thus obtained, so that flow of an electric current having a current density of  $5.4 \text{ mA/cm}^2$  was  
15 observed and a red-light emission having an initial brightness of  $570 \text{ cd/m}^2$  was observed.

In addition, a voltage was applied to this device for 1000 hours with the current density kept at  $5.0 \text{ mA/cm}^2$  under a nitrogen atmosphere. As a  
20 result, the initial brightness was  $530 \text{ cd/m}^2$  and the brightness after 1000 hours was  $480 \text{ cd/m}^2$ . Thus, deterioration in brightness was very small.

(EXAMPLE 4)

A device having a structure as shown in Figure  
25 2 was produced.

A hole-transporting layer 5 was formed by making TPD into a thin film having a thickness of 70

nm on a transparent conductive supporting substrate similar to that in Example 1 by means of vacuum vapor deposition method. Subsequently, an electron-transporting layer 6 was formed by making the exemplified compound No. 1 and aluminum trisquinolinol (weight ratio, 1:50) into a thin film having a thickness of 20 nm by means of vacuum vapor deposition method. The film formation was performed under the conditions where the degree of vacuum at the time of the vapor deposition was  $1.0 \times 10^{-4}$  Pa, and the film-forming rate was 0.2 to 0.3 nm per second.

Next, a cathode 4 was formed on the above-mentioned organic layer in the same manner as in Example 1.

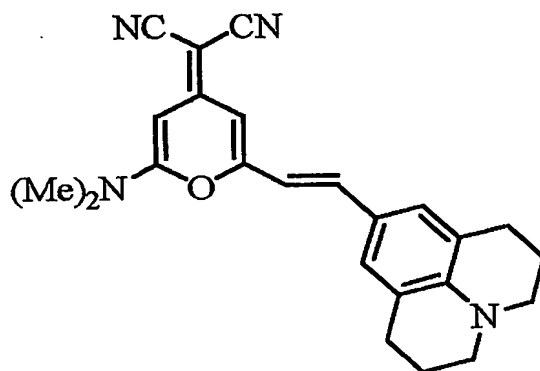
Using an ITO electrode as a positive electrode and an Al-Li electrode as a negative electrode, a direct current voltage of 8 V was applied to the device thus obtained, so that flow of an electric current having a current density of  $5.4 \text{ mA/cm}^2$  was observed and a red-light emission having an brightness of  $26500 \text{ cd/m}^2$  was observed.

In addition, a voltage was applied thereto for 100 hours with the current density kept at  $5.0 \text{ mA/cm}^2$  under a nitrogen atmosphere. As a result, the initial brightness was  $12500 \text{ cd/m}^2$  and the brightness after 100 hours was  $9600 \text{ cd/m}^2$ . Thus, deterioration

in brightness was very small.

(COMPARATIVE EXAMPLE 1)

A device was produced in a similar manner as in Example 4 except that the comparative compound  
5 represented by the following structural formula (US Patent No. 5935720) was used as the compound for the electron-transporting layer.



Using an ITO electrode as a positive electrode  
10 and an Al-Li electrode as a negative electrode, a direct current voltage of 8 V was applied to the device thus obtained, so that flow of an electric current having a current density of 3.7 mA/cm<sup>2</sup> was observed and a red-light emission of 15000 cd/m<sup>2</sup> was  
15 observed.

In addition, a voltage was applied to this device for 1000 hours with the current density kept at 5.0 mA/cm<sup>2</sup> under a nitrogen atmosphere. As a result, the initial brightness was 60 cd/m<sup>2</sup> and light  
20 emission was not detected after 1000 hours.



## INDUSTRIAL APPLICABILITY

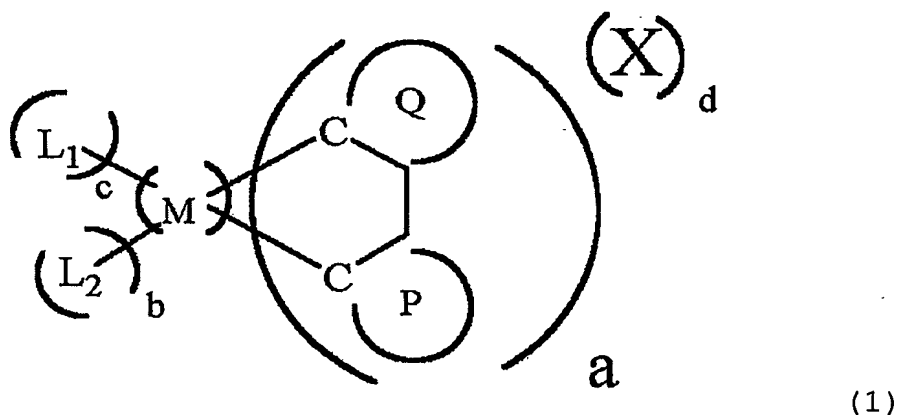
As shown in Examples and Comparative Examples, the organic light-emitting devices using the compounds represented by the general formula (1) of  
5 the present invention exhibit light emissions with a high brightness in spite of a low applied voltage, and the devices are also excellent in durability.

In particular, an organic layer containing a compound represented by the general formula (1) is  
10 useful as an electron-transporting layer and is also useful as a light-emitting layer that emits pure red light.

Moreover, the devices can be produced by the use of vacuum vapor deposition method, casting method  
15 or the like. Large-area devices, therefore, can be produced easily at a relatively low cost.

## CLAIMS

1. An organic light-emitting device comprising  
 a pair of electrodes comprising an anode and a  
 5 cathode, and one or more layers containing an organic  
 compound, the layers being sandwiched between the  
 pair of the electrodes, wherein at least one layer of  
 the layers containing an organic compound contains a  
 compound represented by the following general formula  
 10 (1)



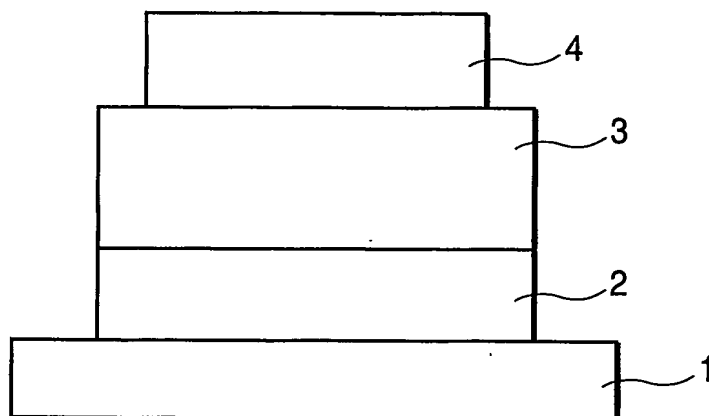
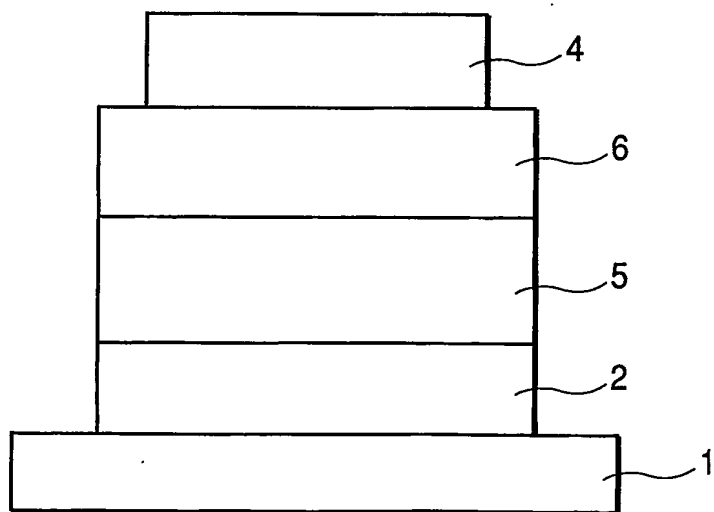
- (In the formula, M is a metal atom, P and Q each are  
 a substituted or unsubstituted aromatic hydrocarbon  
 group or a substituted or unsubstituted aromatic  
 15 heterocyclic group, and P and Q constitute a chelate  
 ligand; P and Q may form a condensed ring with each  
 other; L<sub>1</sub> and L<sub>2</sub> each are a ligand or an oxygen atom  
 that binds to M by a double bond; and L<sub>1</sub> and L<sub>2</sub> may  
 form a condensed ring with each other to become a  
 20 two-coordinate chelate ligand; X represents a counter

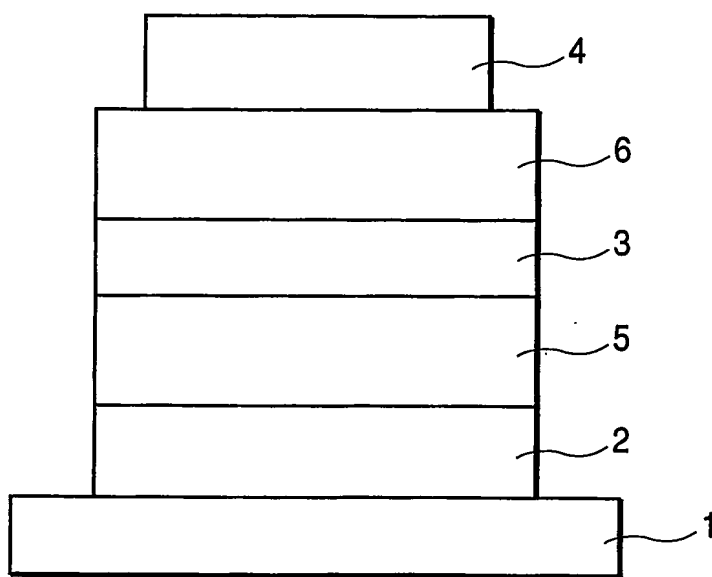
ion of an anion or a cation; a is an integer of 1 to 3; b and c each are an integer of 0 to 4; and d is an integer of 0 to 3, provided that P, Q, L<sub>1</sub>, L<sub>2</sub>, and X each may be the same or different if there are more  
5 than one of these).

2. The organic light-emitting device according to claim 1, wherein the metal atom M is an iridium atom or a platinum atom.

10

3. The organic light-emitting device according to claim 1, wherein the metal atom M is a five-coordinate 16-electron bonding system or a six-coordinate 18-electron bonding system with L<sub>1</sub> and L<sub>2</sub>  
15 and with a chelate ligand comprising P and Q.

*FIG. 1**FIG. 2*

*FIG. 3*

## INTERNATIONAL SEARCH REPORT

International application No.

**PCT/JP03/04483**

**A. CLASSIFICATION OF SUBJECT MATTER**  
 Int.Cl<sup>7</sup> C09K11/06, H05B33/14, H05B33/22

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
 Int.Cl<sup>7</sup> C09K11/06, H05B33/14, H05B33/22

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
<b>X</b>	<b>EP 1191614 A2 (CANON KABUSHIKI KAISHA) 2002.03 27&amp;JP 2002-175884 A</b>	<b>1, 2</b>

☐ Further documents are listed in the continuation of Box C.

☐ See patent family annex.

\* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

**07.07.03**

Date of mailing of the international search report

**22.07.03**

Name and mailing address of the ISA/JP

**Japan Patent Office**

3-4-3, Kasumigaseki, Chiyoda-ku, Tokyo 100-8915, Japan

Authorized officer

**YOKO WATANABE**

Telephone No. +81-3-3581-1101 Ext. 3483



**4V 9279**

# INTERNATIONAL SEARCH REPORT

International application No.  
**PCT/JP03/04483**

## Box I Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
  
2. ☒ Claims Nos.: 3  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:  
The embodiments of the invention described on page 12 do not fall within the scope of the claim. This inconsistency between the claim and the description leads to doubt concerning the matter for which protection is sought, thereby rendering the claims unclear (Article 6 PCT).
  
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
  
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
  
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
  
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.